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Study on a creation of highly ionized cold plasma at atmospheric pressure in the air

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Abstract. This article analyzes the existing ways of creating cold plasma at atmospheric pressure directly into atmospheric air. Research areas listed turns out to be very valuable from a practical point of view, since allows you to create in the plasma gas enough intense ultraviolet radiation, as well as high concentration of physically and biochemically active particles (metastable atoms and molecules, radicals, ozone and other reactive compounds) relatively small specific energy costs. Further in the article, a theoretical comparative analysis of the processes of ozone electrosynthesis in electric sinusoidal and pulsed electric fields is given. The revealed advantage of pulsed electric fields is confirmed by the results of experimental studies

1. Introduction

The object of this article is a low-temperature highly ionized equilibrium plasma created at atmospheric pressure directly in atmospheric air (hereinafter, this plasma will be called "cold"). Such a plasma in dense gases can be called a relatively new, but very promising object in plasma physics. Each of the terms denoting cold plasma has a specific meaning. Thus, low-temperature means that the temperature of the heavy component of such a plasma (ions, neutral particles), as a rule, does not exceed several tens of degrees Celsius, and sometimes is close to room temperature.

Strong ionization means that the cold plasma has a high degree of ionization, in which the number of charged particles leads to the phenomenon of air superconductivity at atmospheric pressure. The specified property of cold plasma is very valuable from a practical point of view, since it allows creating sufficiently intense ultraviolet radiation in the plasma-forming gas, as well as a high concentration of physically and biochemically active particles (metastable atoms and molecules, radicals, ozone and other reactive compounds) with relatively low specific energy consumption.

At present, with the help of cold plasma at atmospheric pressure, it is possible to solve many practical problems that previously seemed unsolvable. Moreover, the use of cold plasma at atmospheric pressure instead of plasma at low pressure makes it possible to get rid of expensive vacuum technology. On the one hand, this significantly reduces the cost of plasma-chemical technologies, and, on the other hand, it makes it possible to carry out a continuous technological process instead of a discrete one at low pressure.

When using low-pressure plasma, it is necessary to load the processed objects into a vacuum chamber, evacuate the chamber, inlet the plasma-forming gas and plasma treatment, after which it is necessary to open the chamber again, unload the processed objects, and then repeat everything. It can even be argued that it is the approaches based on the use of cold plasma in dense gases that determine modern progress in many areas of science, technology, biomedicine and, in particular, in the field of chemical technology.



For the above reasons, much attention is paid to scientific and applied research of cold plasma, numerous conferences are held on this topic, and a huge number of articles and monographs have been published, which cannot be listed within the framework of this article. Let us point out only the most significant from our point of view. For example, a large collection of information on cold plasmas in air at atmospheric pressure is collected in the monograph "Non-equilibrium air plasmas at atmospheric pressure" [1]. The results obtained with the use of cold plasma in solving environmental problems (air pollution by exhaust industrial gas flows) can be found in books [2, 3], as well as in articles [4-6].

A large amount of information about the possibilities of cold plasma in the processing of polymer and textile materials is contained in the collective monograph "Plasma Technology for Hyperfunctional Surfaces" [7], as well as in publications [8–11]. Achievements in the field of biomedicine based on the use of cold plasma are described in the monograph "Plasma for Bio-Decontamination, Medicine and Food Security" [12], as well as in numerous original articles [13,14]. Some prospects arising from the use of cold plasma for the activation of liquids are described in reviews [16, 17] and articles [18, 19]. In addition, the great importance attached to cold plasma by the scientific community is evidenced by such facts as the creation in 2012 of a specialized journal "Plasma medicine" and the formation in 2016 of a consortium of European experts in the field of electric discharge plasma in contact with liquid. In the proposed article, due to the limited scope of its volume, only methods for creating cold plasma in dense gases are considered, a brief overview of the physical and chemical features of the low-temperature, highly ionized equilibrium plasma created at atmospheric pressure directly in atmospheric air is given (hereinafter, this plasma will be called "cold "). In addition, specific examples of gas-phase reactions of active particles created by plasma with organic and inorganic harmful impurities contained in exhaust industrial air flows are given, as well as the results of experiments on the efficiency of destruction of harmful impurities by cold plasma. The content of the review is based on information from sources cited in the bibliography. Since the full title of the monograph or article is given when citing, it is easy to understand which section of the review a particular reference belongs to.

2. Materials and Methods

When the ozone generator is powered from a sinusoidal EMF source, the current i in the circuit has a complex harmonic composition. The average curve of the instantaneous value of the current i contains discontinuities (Figure 1) at the moments of the discharge. The discharge on the period of the supply voltage u occurs twice and stops. The existence of a discharge is possible if the operating voltage $U_{a.c}$ on the ozonizer exceeds a certain minimum voltage U_{cr} . In this case, $U_{a.c}$ is chosen to be less than the voltage of the total electrical breakdown of the discharge gap. When powered by a sinusoidal voltage, the dielectric barrier is heated due to high-frequency discharges during the achievement of a sinusoidal voltage, both positive and negative polarity (Figure 2), the amplitude value, which leads to a decrease in the ozone output. The frequency of these oscillations lies in the range of 300-500 kHz (Figure 3). Therefore, in ozone generators, the electrode covered with the barrier is cooled by running water.

Let us analyze the process of electrosynthesis of ozone when fed with a sinusoidal voltage using the well-known theory of sinusoidal currents with the substitution of parameters corresponding to the barrier discharge process. The total power of the ozone generator (GO) power supply circuit is equal to:

$$P_{gen} = \left(\frac{1}{T} \int_0^T u^2 dt \right)^{0.5} \times \left(\frac{1}{T} \int_0^T i^2 dt \right)^{0.5} \quad (1)$$

Circuit power during barrier discharge

$$P_{pas} = \left(\frac{1}{T} \int_{t_1}^{t_2} u^2 dt \right)^{0.5} \times \left(\frac{1}{T} \int_{t_1}^{t_2} i^2 dt \right)^{0.5} \quad (2)$$

In equations (1) and (2)

$$u = U_{a.c} \sin(\omega_c t - 0.5) \quad (3)$$

$$i = I_{a.c} \sin \omega_c t \quad (4)$$

where ω_c - is the angular frequency of the sinusoidal voltage;

u, i - are instantaneous values of current and voltage;

$U_{a.c.}$ - the amplitude of the sinusoidal voltage;

$I_{a.c.}$ - is the amplitude of the discharge current;

T - is the period of the sinusoidal voltage;

t_1 - is the start time of the discharge;

t_2 - is the discharge completion time.

The power loss is

$$P_{loss} = P_{gen} - P_{disch} \quad (5)$$

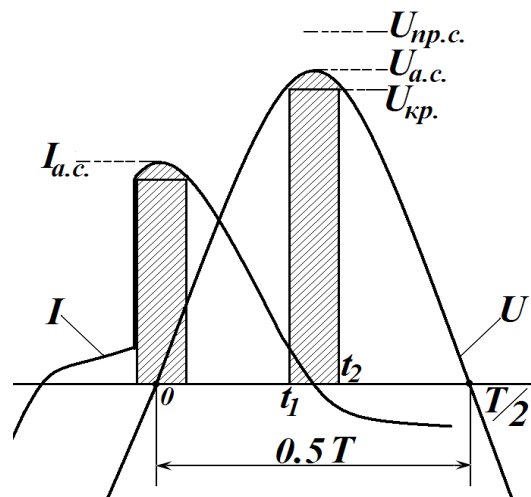


Figure 1. Changes in voltage and current on the ozonizer when powered by a sinusoidal voltage

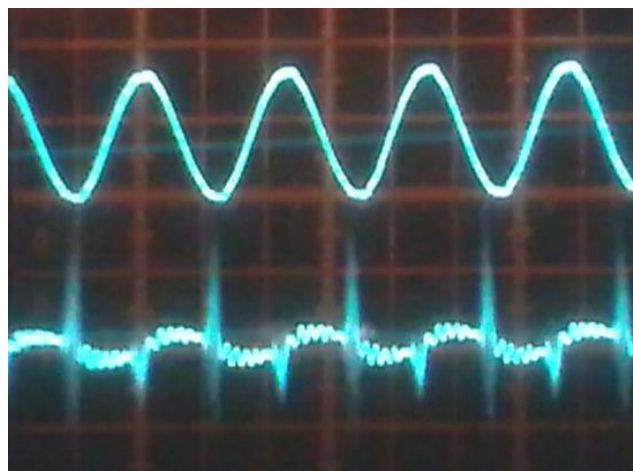


Figure 2. Oscillograms of voltage and current changes on the ozonizer when powered by a sinusoidal voltage

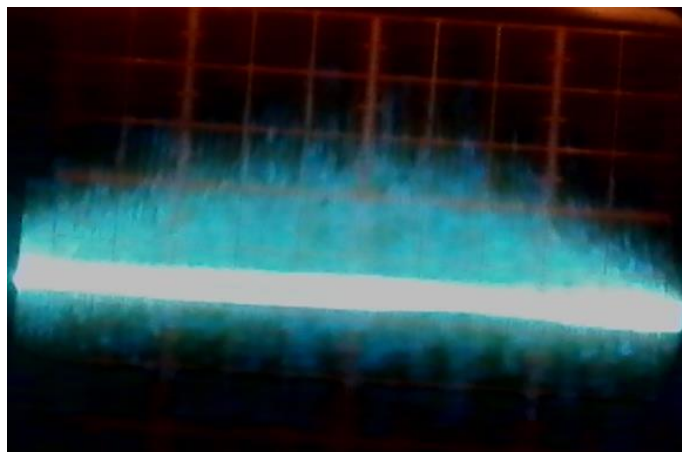


Figure 3. The oscillogram of the discharge current at the moments when the sinusoidal voltage reaches the amplitude value

From the analysis of formulas (1 ... 5) it follows that a significant power loss falls on the time at which there is no discharge in the GO. This is confirmed by the oscillograms of the sinusoidal voltage and discharge current presented in Figures 2 and 3.

From the analysis of discharge processes in the circuit “secondary winding of a step-up transformer - ozone generator”, when powered by a sinusoidal voltage, the discharge currents are periodic, namely, they are determined by the applied sinusoidal periodic voltage.

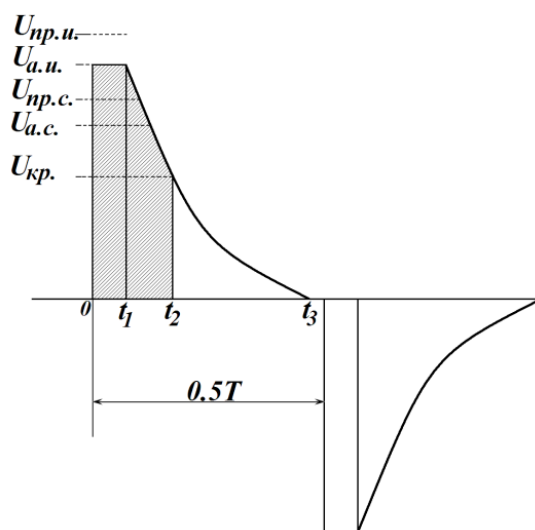


Figure 4. Voltage on the ozonizer when powered by periodic voltage pulses with a crest factor of more than 5

From this follows a working hypothesis: the efficiency of the ozone electrosynthesis process can be increased by using periodic voltage pulses that have a shape similar to the shaded area of a sinusoidal voltage, i.e. rectangular with a large crest factor (Figure 4). In this case, it is possible to increase the amplitude of the supply voltage $U_{i.a}$ above the breakdown threshold of the sinusoidal voltage $U_{p.r.s}$, which is characterized by the overvoltage coefficient.

$$K = U_{u.a} / U_{np.c}. \quad (6)$$

With pulsed power, the GO capacitance is charged to the amplitude value of the voltage, which is higher than the amplitude of the sinusoidal voltage. In the time interval 0 - t1 (Fig. 4), the discharge power is determined by the dependence

$$P_{0-t1} = I_{a.u.} \cdot U_{a.u.} \quad (7)$$

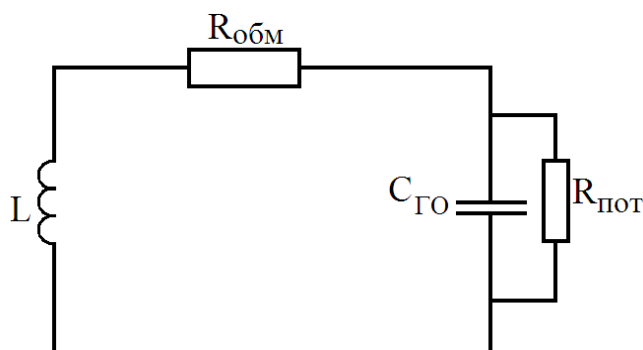


Figure 5. Equivalent circuit of the discharge circuit "Secondary winding of step-up transformer - ozone generator"

In the pause between pulses (time from t1 to t2), the discharge power will be determined by the charge accumulated in the capacitance of the GO, which will be discharged on the loss resistance Rpot of the GO, the resistance Robm and the inductance L of the secondary winding of the step-up transformer, which are elements of the oscillatory circuit (Figure 5). At the same time, due to the fact that $R / 2L < (CL) 0.5$, we have low attenuation in the circuit, which is the sum of the loss currents on Rpot (the first term on the right side of the equation) and the process in the circuit $R_{обм} LC_{zo}$:

$$P_{t1-t2} = \frac{U_{a.u.}^2}{R_{nom}} e^{-\frac{2t}{R_{nom}C_{ГО}}} + \frac{U_{a.u.}^2}{R_{обм}} e^{-\frac{tR_{обм}}{2L}} \left(\cos \omega_{к.к} t + \frac{R_{обм}}{2\omega_{к.к}} \sin \omega_{к.к} t \right), \quad (8)$$

where $\omega_{к.к}$ - is the angular frequency of the oscillatory circuit.

Starting from time t2, the voltage of the discharge gap decreases below the critical Ucr, the discharge stops, and the capacitance is discharged by Rобм, which is the power loss.

Analysis of equations (7-9) shows that the main part of the energy of periodic voltage pulses is spent on the discharge process, which is accompanied by ozone electrosynthesis.



Figure 6. Oscillograms of voltage and current changes in the discharge circuit when supplied with pulsed voltage

In high voltage ozone generator power circuits, the active resistance of the secondary winding of the step-up transformer can be in the range from 2 to 20 kOhm, inductance from 50 to 200 H, capacitance of ozone generators from 10^{-7} to 10^{-9} F. In this case, the condition $(R_{obm} / 2L) < (LC)$ and the discharge in the circuit proceeds with low attenuation.

According to the shape of the electrodes, tubular and plate ozonizers are distinguished. For the operation of ozonizers, this difference is not essential, although in the latter case it is more difficult to avoid some edge effects.

The most widely used tubular ozonizers, which consist of a package of tubular electrodes placed in a common cylindrical housing. Proceeding from this, let us consider the change in the electric field strength in the discharge gas gap and in the dielectric barrier of the tubular ozonizer.

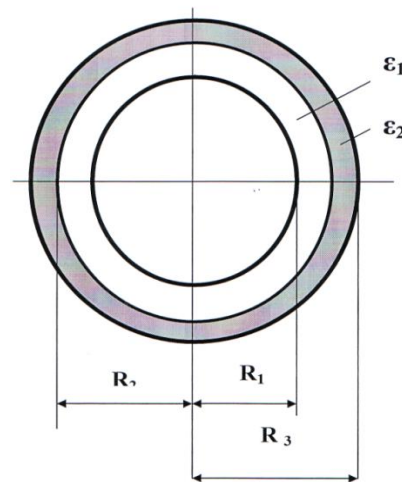


Figure 7. Main parameters of tubular ozone generator

A tubular ozonator is a two-layer capacitor, in which the electric field is created between two cylindrical surfaces with a common axis (Figure 7) and has a radial direction. Due to symmetry, equipotential surfaces have a cylindrical shape; the axes of these surfaces coincide with the common axis of the electrodes, and, of course, at all points of the same equipotential surface, the magnitude of the tension is constant and decreases from one equipotential surface to another.

In this case, the capacity of the gas gap will be equal to:

$$C_1 = 2\pi\ell\varepsilon_1\varepsilon_0 / (\ln R_2/R_1), \quad (9)$$

where ℓ - is the length of the capacitor, m;

$\varepsilon_0 = 8.85 \cdot 10^{-12}$ F/m – electrical constant.

Dielectric barrier capacitance

$$C_2 = 2\pi\ell\varepsilon_2\varepsilon_0 / (\ln R_3/R_2). \quad (10)$$

Capacitance of capacitors connected in series

$$C = C_1 C_2 / (C_1 + C_2). \quad (11)$$

The electric field strength of a cylindrical capacitor is determined by the formula:

$$E = Q / \varepsilon \varepsilon_0 2\pi R, \quad (12)$$

where R is the radius of the equipotential surface for which the electric field strength is determined.

Since, when capacitors are connected in series, the charges on all plates are the same in magnitude, then

$$Q = C_1 U_1 = C_2 U_2 = CU. \quad (13)$$

From here you can determine the electric field strength in each layer of the capacitor.

Electric field strength in the gas layer

$$E_1 = CU / \epsilon_1 \epsilon_0 2\pi R_{\epsilon_1}, \quad (14)$$

where R_{ϵ_1} - is the radius of the equipotential surface in the gas layer, for which the electric field strength is determined. Electric field strength in the barrier layer is calculated using following equation (15):

$$E_2 = CU / \epsilon_2 \epsilon_0 2\pi R_{\epsilon_2}, \quad (15)$$

where R_{ϵ_2} - is the radius of the equipotential surface in the barrier layer, for which the electric field strength is determined.

3. Results and Discussion

For numerical analysis, we use the results of experimental studies. In the experiments, an ozone generator was used with the following parameters: $R_1=0.011\text{m}$; $R_2=0.017\text{ m}$; $R_3=0.02\text{ m}$; $\epsilon_1=1$; $\epsilon_2=7$ (Figure 7). A grid with $2 \times 2\text{ mm}$ cells was used as a potential electrode, which was fixed on the surface of the glass barrier. The experiments were carried out using a machine generator of periodic acute-angled voltage pulses. For comparison, the parameters obtained using sinusoidal voltages were used (Table 1).

In the experiment for both types of voltage, the effective voltage value was 11.5 kV . In this case, the voltage amplitude was 16.215 kV for the sinusoidal voltage, and 57.5 kV for the pulse voltage. The overvoltage factor was 3.55 . The current of the discharge circuit with an impulse voltage is 20 times greater than with a sinusoidal voltage. Accordingly, the conductivity of the discharge gap increases by the same amount. This leads to the conclusion that under the influence of the amplitude value of the electric field strength of the pulsed voltage both in the dielectric barrier and in the gas gap, a low-temperature plasma is formed, which is characterized by high conductivity, luminescence in the ultraviolet region of the optical radiation spectrum, and the absence of heating of the dielectric barrier. The last two effects are associated with the known theory of the structure of materials, according to which the transition from a high energy zone to a lower one is accompanied by energy absorption and luminescence in the visible region of the optical radiation spectrum (Table 3).

In contrast to the processes of ozone electrosynthesis in an electric field of a sinusoidal voltage, in pulsed electric fields, the discharge process is aperiodic in nature, i.e. the discharge process proceeds due to the energy accumulated in the discharge circuit under the action of a high voltage pulse with an overvoltage.

Table 1. Parameters of sinusoidal and impulse voltages

Parameter name	Sinusoidal 50 Hz	Pulse 250 Hz
Operating voltage at the transformer input, V	80	70
Effective current at the input of the transformer, A	0.32	2.8
Power of the transformer primary winding circuit, W	25.6	196
Rated voltage, kV	11.5	11.5
Crest factor	1.41	5
Voltage amplitude, kV	16.215	57.5
Overvoltage factor		3.55
Discharge current, mA	20	400
Ozone generator circuit power, W	230	4600
Discharge circuit resistance, kOm	575	28.7

To stabilize the discharge current in amplitude, the pulse repetition rate must be chosen from the condition that in the pause between pulses, all processes of recombination and movement of space charges in the discharge gap must be completely completed or reduced to a level at which they cannot prevent the passage of the next voltage pulse of opposite polarity (time t_3 Figure 3). This frequency should be selected taking into account the parameters of the discharge gap circuit: active and reactive

conductance of the secondary winding of the step-up transformer, capacitance and leakage resistance of the ozone generator.

It is obvious that the choice of the optimal frequency of repetition of voltage pulses must take into account the parameters of the step-up transformer and ozone generator in each individual application. The amplitude values of sinusoidal and impulse voltages were used in the calculations. This is due to the fact that the discharge processes occur in a narrow section of voltages adjacent to their amplitudes.

Table 2. The results of calculations of the influence of the parameters of the dielectric barrier on the electrical parameters of the ozone generator

Defined parameter	Designation parameter	Used formula	Parameter value
1. Gas gap capacity, 10^{-11} F	C_1	(10)	5.1
2. Dielectric barrier capacitance, 10^{-10} F	C_2	(11)	9.57
3. Ozone generator capacity, 10^{-11} F	C	(12)	5.03
4. Radius of the equipotential surface in the gas layer, m	R_{e1}	$(R_1+R_2)/2$	0.014
5. Radius of the equipotential surface of the barrier, m	R_{e2}	$(R_3+R_2)/2$	0.0185
6. Electric field strength in the gas layer, kV/m	E_1	(15)	1048* 3724**
7. The intensity of the electrical which field in the barrier, kV/m	E_2	(16)	113* 402**

* - parameters for sinusoidal voltage;

** - parameters for impulse voltage.

The heating process of the dielectric barrier is given in Table 3.

Table 3. The process of heating the dielectric barrier of a tubular ozone generator when powered by a sinusoidal and pulsed voltage

Current frequency	Ozone generator operating time, min			
	0	10	20	30
Sinusoidal, 50 Hz	23.2	35.6	47.5	52.3
Pulse, 250 Hz	23.5	24.4	24.6	24.7

The analysis shows that the presence of a dielectric barrier significantly increases the electric field strength in the gas layer, which contributes to the process of ozone electrosynthesis. The barrier makes it possible to obtain electric field strengths that significantly exceed the electrical strength of gaseous dielectrics, which is the main purpose of the barrier. There is practically no heating of the dielectric barrier.

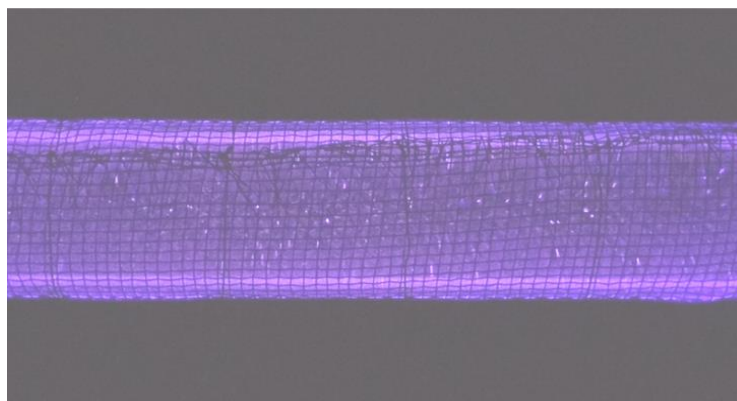


Figure 8. Photograph of the glow of a tubular ozonizer in the ultraviolet spectrum

Theoretically, efficiency of the developed method of powering ozone generators with a pulsed voltage is much higher than the efficiency. process of electrosynthesis of ozone at a sinusoidal voltage.

To stabilize the discharge current, the pulse repetition rate must be chosen from the condition under which all processes of the movement of charges in the discharge gap must be completed before the voltage pulse opposite in sign is applied.

The device of ozone generators can be implemented using plate or tubular (Figure 7) electrodes. When the tubular ozone generator is powered by periodic pulses with a duty cycle of more than 5, the electric field strength is 3.7 times higher than the field strength when powered by a sinusoidal voltage. Figure 8 shows a photograph of the glow of a tubular ozonizer in the ultraviolet spectrum.

Ozone generators use independent electric discharges in which various elementary processes occur: separation, movement and propagation of charged particles. ionization processes take place. Discharge current pulses are randomly distributed in time and are used together with the function of a discrete random process, the implementation of which is random in amplitude and frequency. Numerous observations show that the frequency of current pulses varies from expected to thousands of frequencies (Figure 6). The discharge process falls on a small part of the voltage, which explains the low efficiency. ozone electrosynthesis process at sinusoidal voltage.

The listed properties of a self-sustained discharge lead to the instability of the discharge currents in frequency and amplitude, to the blocking of the discharge, the transition to a spark or arc form. Obviously, by ensuring the stability of the discharge processes, it is possible to increase the efficiency of the ozone electrosynthesis process.

To stabilize the discharge current, it is necessary that in the pause between pulses, all discharge processes be completed or reduced to a degree that does not prevent the flow of a voltage pulse opposite in sign.

The device for electrosynthesis of ozone (Figure 9) consists of a power supply unit 1, a generator of periodic voltage pulses 2 of acute-angled or rectangular shape with a duty cycle of more than 5 (duty cycle is the ratio of the pulse repetition period to its duration), step-up transformer 3, ozone generator 4, voltage divider 5, kilovoltmeter 6, ammeter 7.

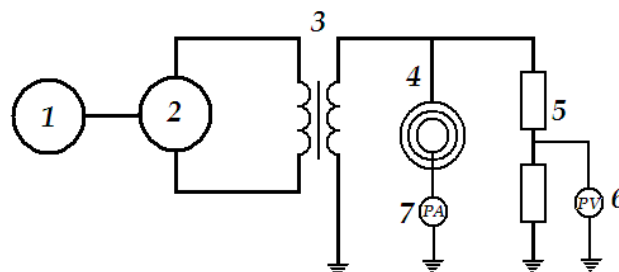


Figure 9. Schematic diagram of the power supply of the ozone generator with a pulsed periodic voltage

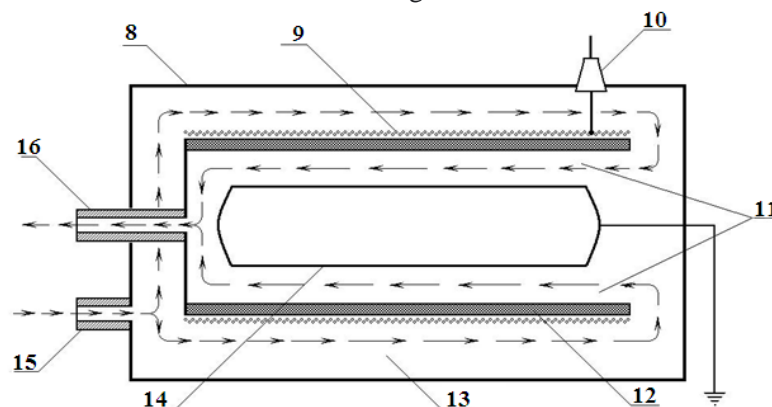


Figure 10. Ozone generator device

The ozone generator consists (Figure 10) of a dielectric housing 8, a potential electrode in the form of a grid with 2x2 mm cells 9, a bushing insulator 10, a low-temperature plasma zone 11, a dielectric barrier made of heat-resistant glass 12, an ultraviolet irradiation zone 13, a grounded cylindrical electrode 14, branch pipe for air supply 15, branch pipe 16 for the outlet of ozone-containing gas. The device works as follows.

From the power source 1, power is supplied to a machine or electronic generator of acute-angled or rectangular periodic voltage pulses 2 with a crest factor of more than 5. Periodic voltage pulses are increased by a transformer 3 and fed to the potential electrode of the ozone generator 4. The voltage supplied to the ozone generator is controlled by a kilovoltmeter 6, which connected through a voltage divider 5. The discharge current is measured by a milliammeter 7.

The amplitude of the voltage applied to the potential electrode is 3.55 times greater than the amplitude of the sinusoidal voltage, which significantly exceeds the electric field strength both in the gas space of the ozone generator and in the dielectric barrier. The magnitude of this intensity is sufficient for the formation of low-temperature plasma. The presence of low-temperature plasma is proved by the presence of luminescence in the ultraviolet region of the spectrum and the absorption of energy. The latter factor is especially important because it eliminates the operation of cooling the dielectric barrier and the need for air preparation before processing.

The process of electrosynthesis of ozone proceeds in two stages. At the first stage, the processed air, through the pipe 15, enters the zone of ultraviolet radiation 13. The ultraviolet radiation enters zone 13 from the zone of low-temperature plasma through a potential electrode made of a grid with 2x2 mm cells and a dielectric barrier 13. Voltage is applied to the potential electrode through the through passage insulator 10.

At the second stage of ozonation, the partially ozonized gas enters the zone of action of the low-temperature plasma 11. The ozonized gas is removed from the ozone generator through the branch pipe 16.

4. Conclusions

1. The use of a pulsed periodic voltage, the amplitude of which is 5 times higher than the amplitude of the sinusoidal voltage, leads to a strong ionization of the gas in the discharge gap, and the formation of a low-temperature plasma.
2. In the low-temperature plasma channel, the conductivity increases sharply, which leads to the formation of large discharge currents and weakly damped oscillations, the attenuation of which is determined only by the resistance of the secondary winding of the step-up transformer, which, together with the capacitance of the discharge gap, form an oscillatory circuit.
3. In the low-temperature plasma channel, under the action of the discharge current, the ozone yield and the efficiency of the generator significantly increase, due to the fact that the oscillatory process proceeds due to the energy accumulated in the oscillatory circuit at the moment of applying a high voltage pulse with overvoltage.
4. The discharge process proceeds due to the transition of charged particles from higher energy zones to low energy zones, and this process is accompanied by plasma glow with a frequency in the UV radiation region and energy absorption.
5. The process of energy absorption makes it possible to exclude the operation of cooling the dielectric barrier from the technology of ozone electrosynthesis.
6. The process of electrosynthesis of ozone proceeds in two stages: at the first stage, ozone is formed in the zone of UV radiation; at the second stage - in the zone of low-temperature plasma.
7. The discharge process in the power supply circuit of the ozone generator is determined by the active resistance of the secondary winding of the step-up transformer, while the gas gap of the ozone generator has practically superconductivity.

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